Preliminary Studies of ATW Multiple Strata Fuel Cycle Performance

R. N. Hill and T. A. Taiwo

Argonne National Laboratory
9700 South Cass Avenue, Argonne, Illinois 60439
BobHill@anl.gov; Taiwo@anl.gov

Abstract - *Two primary goals have been identified for the multiple strata system studies: a) reduce the number of ATW systems required to complete the mission, and b) enhance the performance of the final strata ATW system. For the first strata, both a fast-spectrum advanced liquid metal reactor (LMR) and a thermal-spectrum gas-cooled reactor (GT-MHR) were evaluated; and both nonfertile and fertile fuel forms were considered. For this study, the focus was on fuel cycle options where the LWR transuranics (plutonium and minor actinides) remain mixed to alleviate proliferation concerns. Fuel cycle analyses were conducted for once through and single recycle fuel management in the first strata reactor system. Preliminary results show that the multiple strata options realize the primary goal of partial TRU consumption prior to the ATW campaign. In both systems, ~30% of the TRU mass was consumed in the once-through first strata irradiation. However, it was shown to be quite difficult to achieve improved ATW performance; only small impacts on the burnup swing (1-2%Δk) were observed with significant penalties (~25%) in ATW discharge burnup.*

I. INTRODUCTION

Previous work in the Accelerator Transmutation of Waste (ATW) program focused on direct utilization of the transuranics (TRU) contained in LWR spent fuel. As the ATW was the sole device being utilized to destroy the hazardous material, this base fuel cycle will be denoted single strata. Last year, an alternative fuel cycle strategy was proposed for the gas-cooled ATW option.[1] In this case, the TRU material was first irradiated in a thermal-spectrum region (to burn the plutonium) with subsequent irradiation in a fast spectrum region (to burn the minor actinides). This scheme will be denoted double strata as the material is transmuted in two distinct environments. In principal, one could envision fuel-cycle schemes with multiple irradiation conditions; in addition, one could obtain these variations within different regions of the same machine (as planned in the thermal/fast concept) or in several distinct devices (e.g., LWR MOX followed by ATW). An unlimited number of multiple strata fuel cycles can be conceptually conceived; and these strategies could employ a diverse set of neutron source and fuel processing technologies making consistent evaluation of the options difficult.

The primary goal of the ATW system remains – to transmute the hazardous components of spent nuclear fuel. Thus, the majority of the TRU material needs to be fissioned, producing ~1 MW-day of energy for every gram. However, other systems may be more efficient in consuming this material, particularly early in the process when the fissile content is still high. The general perception is that the ATW system will be required for the ultimate destruction of the most hazardous species (minor actinides), but that ATW will also be more costly than reactor systems. Therefore, it may be desirable to achieve partial burning in reactor systems, limiting the number of ATW systems required to finish the mission. As a result, two general goals were identified for the multiple strata system studies: a) reduce the number of ATW systems required to complete the mission, and b) enhance the performance of the final strata ATW system.

One key design issue is the choice of nonfertile fuel forms or more conventional (uranium-based) fuel forms for the first-strata reactor system. Nonfertile fuel forms are appropriate for ATW where the primary goal is to destroy the material as quickly as possible. However, such fuel forms may cause safety and/or performance problems if they are employed in power producing reactors. Thus, it may be preferable to slow down the net destruction rate but utilize more conventional fuel forms. Therefore, both fertile and nonfertile fuel forms were considered in these multiple strata system studies.

Another key issue is the degree of transuranic (TRU) separation in the initial LWR spent fuel processing step. Preliminary studies by LANL focused on a double strata fuel cycle strategy that significantly consumes plutonium in a LWR (using nonfertile fuel) followed by final disposal in a ATW system.[2] In this scenario, a more complete separation of the TRU (into plutonium and minor actinide) is performed in the initial LWR spent fuel processing. Separation of the plutonium from the minor actinides improves ATW system performance by burning the plutonium in a separate system and exploiting the fertile properties of the minor actinides. However, there are major proliferation concerns associated with fuel cycle strategies utilizing more detailed separation of the TRU; the standard PUREX process has been rejected as a recycle technology in the U.S. because of the presence of a pure plutonium stream. The proliferation resistance of alternative processing techniques (e.g., electrometallurgical) derives primarily from the fact that the minor actinides are retained with the plutonium, significantly reducing its attractiveness. Thus, mixed (non-separated) LWR TRU fuel cycles are preferred, and are the focus of this study.

An overview of the multiple strata approach evaluated in this work is shown in Fig. 1. Initial system studies will consider dry-processing options in which the LWR

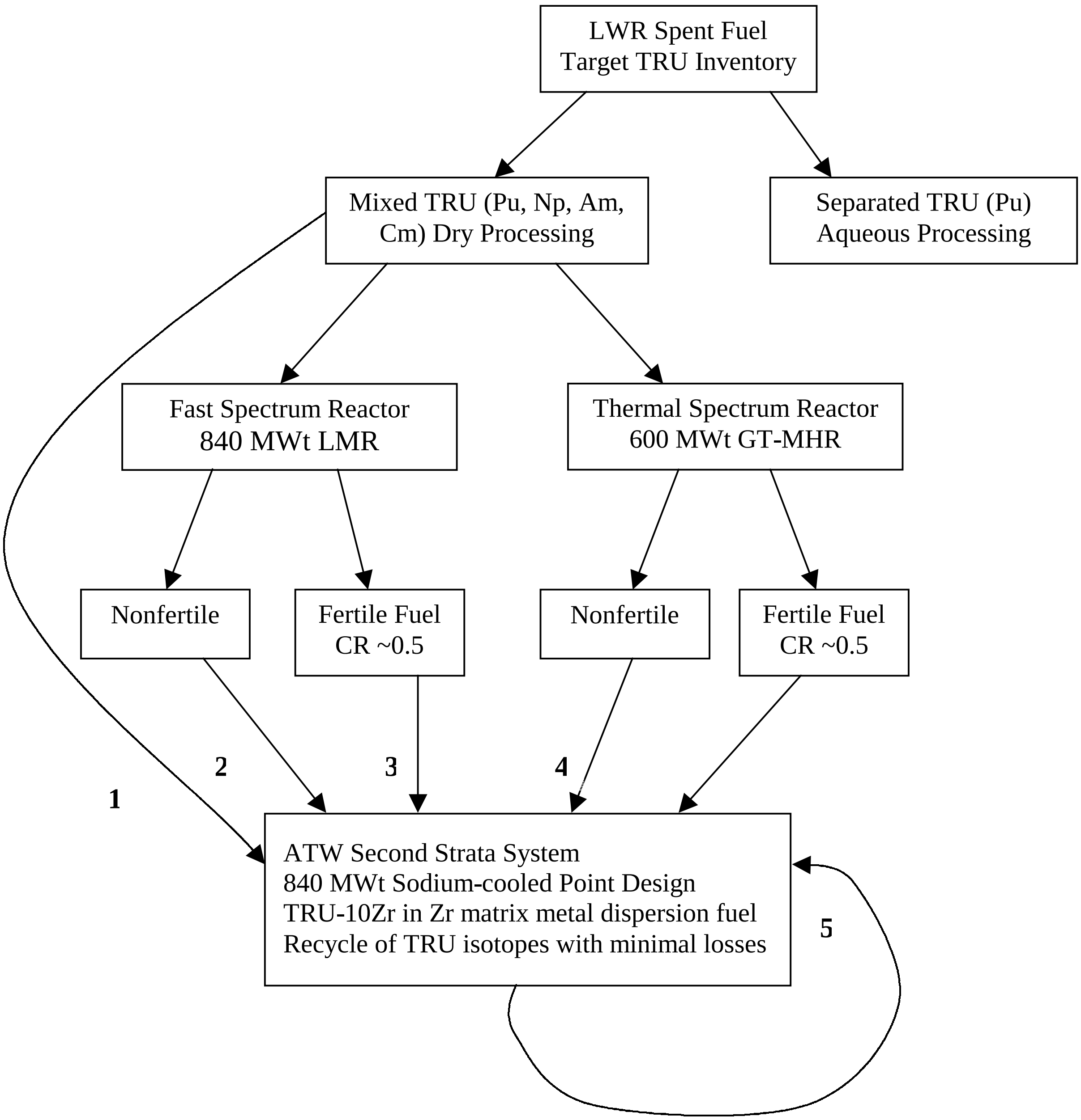

Fig. 1. Flow Sheet for Preliminary Multiple Strata Studies.

transuranics (plutonium and minor actinides) remain mixed. A wide variety of reactor technologies (e.g., existing or advanced LWRs, gas-cooled reactors, liquid-metal cooled fast reactors) could be employed for the initial burning of the TRU material. Regarding TRU consumption, the key parameters are a) irradiation environment (fast or thermal spectrum), b) discharge burnup of first strata fuel form (extent of burning), and c) presence of fertile material (production of new TRUs). As shown in Fig. 1 both fast and thermal systems using nonfertile and fertile fuel loading, are considered. Furthermore, the initial work was limited to a single pass through the first strata reactor.

In other studies, irradiation in the first strata was extended to a single recycle strategy. In those studies, it is assumed that fuel discharged from the once-through scheme is processed to extract the heavy metals (again without separation of the TRUs), fabricated into fuel for the first strata systems, and irradiated a second time in the first strata reactor. This allows additional consumption of the TRU inventory before introduction into the second strata ATW system.

The computational methods employed in this study are briefly identified in Section II. The first-strata fast and thermal reactor systems are described in Sections III.A and III.B, respectively, and results for first-strata system performance are summarized. In Section IV, the impact of first strata irradiation on performance of the second strata ATW system is evaluated. Finally, the key conclusions from these preliminary studies are summarized in Section V.

II. COMPUTATIONAL METHODS

For the fast reactor systems and the fast-spectrum ATW system, reactor and fuel cycle performance was evaluated using the REBUS-3 fuel cycle analysis code.[3] For each external feed composition (e.g., LWR spent fuel TRU in the first strata, first strata discharge in the second strata), the TRU mass loading in the fuel was determined using the REBUS-3 enrichment search techniques for an EOC multiplication factor constraint. The region-dependent multigroup cross sections are based on ENDF/B-V.2 basic data processed using the MC^2-2 and SDX codes[4,5] for a 21-group energy structure. The computational techniques employed in this study are described in more detail in Refs. 6 and 7.

The REBUS-3 fuel cycle analysis code was also used for evaluating the core performance of the first-strata, gas-cooled system. Burnup-dependent, 23-group cross sections, generated using the DRAGON lattice code,[8] were employed in the REBUS-3 calculations. The 69-group cross-section library used in the DRAGON calculations is based on ENDF/B-VI nuclear data.

III. FIRST STRATA REACTOR SYSTEM RESULTS

In this section, the first-strata reactor systems are briefly described and their performance results for the envisioned scenarios are summarized.

III.A. Fast Spectrum Reactor

For the fast-reactor first-strata system, the 840 MWt Advanced Liquid Metal Reactor (ALMR) design was utilized for these preliminary studies. This design was developed in the former U.S. fast reactor program by General Electric and Argonne National Laboratory in the 1985-1995 time frame; conventional and burner configurations for the weapons plutonium disposition mission are described in detail in Ref. 9. First, the nonfertile-fuel fast-reactor concept is described. For this design, the core volume was maximized by using a large number of fuel assemblies and a tall core (active height of 42 inches). This configuration leads to a large core TRU inventory that tends to reduce the reactivity loss rate. Note that low reactivity losses are quite important for reactor performance since criticality must be maintained; excess reactivity must be introduced and control systems must compensate all reactivity losses. To further increase the reactor TRU inventory and to re-introduce some Doppler reactivity feedback, a fixed poison (hafnium, in this case) was also introduced. The fuel form described in Ref. 9 is a TRU/Zr alloy cast into a Hf/Zr sheath with the sheathed slug encased in standard size fuel pins. For this study, calculations were also performed using the proposed ATW fuel form using TRU/Zr fuel particles in a Zr dispersion matrix within the same fuel pins and assembly design.

The conventional burner design in Ref. 9 utilizes standard fertile fuel – ternary metal U/TRU-10Zr alloy with maximum TRU content of ~30%. The core configuration is identical to the pure burner (nonfertile) design. However, the core geometry is spoiled by reducing the core height to 18 inches. This increases the leakage and reduces the conversion ratio (CR) to ~0.5; this allows net consumption of the TRU feed at roughly half the rate of the pure burner concept.

Reactor performance was evaluated for both of these systems, using a feed composition based on processed LWR spent fuel transuranics. Key reactor performance parameters for the fast-reactor first strata options are summarized in Table I. Each case operates on a one-year cycle length with either 75% or 85% capacity factor. The high inventory pure burner cases require very long fuel residence time, 10 to 14 years, to achieve their full (fluence-limited) burnup, as compared to 7 years for the conventional burner design. As expected, the net TRU consumption rate of the conventional burner (CR of 0.5) is roughly half the pure burner destruction rate. Large variations in the TRU inventory are observed: 3.9 MT for the reduced volume fertile system, 5.3 MT with the ATW fuel pure burner, and 7.9 MT with the fixed poison added.

Even with high TRU inventory, the reactivity loss rate of the pure burner systems is significantly larger than the

TABLE I

Fast Reactor First Strata System Performance

ALMR Burner Configuration		Nonfertile Hf Sheath	Nonfertile Dispersion	Dispersion *Recycle*	Fertile Fuel	Fertile *Recycle*
Number of fuel batches		14	10	10	7	7
Cycle irradiation time (days)		274	274	274	310	310
Conversion Ratio		0	0	0	0.51	0.49
Net TRU Consumption rate (kg/y)		233	234	234	116	120
Fuel Enrichment (TRU/HM or TRU/matrix vol.)	Inner zone	36.6	19.6	22.5	25.9	26.7
	Outer zone	45.3	23.8	27.3	32.3	33.4
BOEC Inventory (kg)	TRU	7875	5288	6248	3874	--
	Total HM	7875	5288	--	13914	13916
Burnup reactivity loss (%Δk)		3.08	4.89	4.14	2.51	2.45
Peak linear power (W/cm)		145	143	137	265	265
Discharge burnup	MWd/kgHM	320	339	295	118	118
	Atom %	34.0	35.9	31.3	12.5	12.5
Peak fast fluence (10^{23} n/cm^2)		3.83	3.98	4.02	3.77	3.77

fertile fuel design; the fixed Hf poison reduces the burnup swing by ~2%Δk. The discharge burnup of the nonfertile fuel, ~34 atom%, is similar to that of the ATW system point design; whereas, the fertile-based fuel only achieves ~10 atom% burnup of the uranium-dominated heavy metal. Thus, additional processing will be required in the fertile fuel case. In addition, a later step to remove the uranium will be required (before introduction into ATW); however, the initial LWR spent fuel separation may be simplified since complete uranium removal is not required.

For the dispersion fuel and fertile fuel options, irradiation in the first strata was extended to a single recycle strategy, also shown in Table I. The results indicate that the non-fertile option destroys ~35% of the TRUs in the once-through scheme; TRU is destroyed at roughly half this rate in the fertile system. An additional ~30% destruction is obtained in the second pass (recycle stage) of the non-fertile design. Note that because of the initial burning of fuel in the startup stage (once-through case), the fissile content of the fuel for the recycle stage is lower than that for the once-through case, this leads to a higher fuel enrichment (or volume fraction) in the recycle cases. The higher fuel volume fraction implies a higher initial fuel mass, which results in a lower discharge burnup and reactivity loss for the recycle stage, since the cycle length is held constant. These performance effects are much more pronounced in the non-fertile fuel case where the isotopic changes from the once-through irradiation are more pronounced (compositions given in more detail in Section IV).

III.B. Thermal Spectrum Reactor

For this study, a 600 MWt critical gas-cooled reactor (GT-MHR) was considered. The configuration replaces the interior fast zone of a typical GT-MHR system[10] with graphite reflectors, and maintains the three ring annular core with a three batch scattered refueling pattern. This system operates with nonfertile TRISO fuel particles and employed erbium oxide burnable poison; fertile fuel utilization in the thermal systems was not considered. The first strata performance of the GT-MHR is summarized in Table II. A net consumption of 69% of the Pu-239, but only 31% of the

TABLE II

Thermal Reactor First Strata System Performance

Parameter		Nonfertile Once-Through	Nonfertile *Recycle*
Batch Initial Heavy Metal Loading, kg		351.3	351.3
Batch Initial Erbium-167 Loading, kg		0.21	0.21
Number of Fuel Batches		3	3
Cycle Length, Days		180	100
k_{eff}	Initial	1.032	1.025
	Final	1.000	0.994
Consumption levels			
Pu-239 Consumption, %		69	52
Net Plutonium Consumption, %		34	19
Net Heavy Metal Consumption, %		31	18

TRU is achieved during a single irradiation campaign (three cycles). Preliminary studies showed that the nonfertile thermal spectrum system is quite sensitive to changes in the feed isotopics. In particular, with an increase in decay time for the LWR feed from 10 years to 25 years (more Pu-241 decay), the achievable TRU net consumption in a single pass decreases from 46% to 31%.

In a similar manner to the fast reactor studies in Section III.A, a single recycle in the first strata was also evaluated. In this analysis, the initial fuel and poison loading are kept constant. Thus, the impact of the lower fissile content in the recycle stage of the GT-MHR design is to reduce the cycle length from 180 to 100 days as shown in Table II. The lower cycle length results in a significant reduction of the heavy metal consumption from ~30% to ~20%. This result again exemplifies that the thermal system is quite sensitive to isotopic variations; the changes in a fast spectrum system with recycle are much smaller as shown in Table II.

IV. SECOND STRATA ATW SYSTEM RESULTS

In this section, the impact of the first strata irradiation on the second strata ATW system is evaluated. Imposition of the first strata reactor between the initial LWR material and the ATW changes the composition of the ATW feed stream. The isotopics of the different feed streams shown in Fig. 1 are compared in Table III. In general, the thermal-spectrum system rapidly destroys Pu-239, but builds in Pu-241 at this burnup level. This eliminates the primary fissile species but will have a negative impact on the radiotoxicity of the spent fuel because the Np-237 chain (Pu-241 decays to Am-241 which decays to Np-237) fraction increases significantly (from 18 to 30%), and Np-237 is the key long-lived TRU isotope for repository considerations. Conversely, the fertile fuel system roughly conserves the Pu-239 fraction; this limits the production of higher actinides but retains the fissile content of the TRU. Results in Table III also indicate that the fissile plutonium content of the diverse first strata scenarios are bracketed by the LWR spent fuel (57% fissile plutonium) and ATW recycle (25% fissile plutonium) streams.

Finally, the impact on ATW performance of the feed variations shown in Table III was evaluated. For this study, the 840 MWt sodium-cooled ATW system point design[6] was used. This design utilizes the TRU-10Zr in Zr matrix dispersion fuel and was optimized for the LWR TRU discharge case (feed #1). Using REBUS-3, the startup core performance was evaluated for each feed composition. An enrichment search was performed to determine the enrichment required to achieve a BOC multiplication factor of 0.97. The fuel reload strategy was retained with a cycle length of six months with a 85% capacity factor; the inner core uses 7 batches with 8 batches for the lower enrichment outer core region. For these scoping studies, only eigenvalue computations were performed; previous work has shown that this method is adequate for performance predictions,

TABLE III

Comparison of Potential Feed Streams (Isotopic Mass %)

Nuclide	1-LWR Discharge	2-LMR Nonfertile		3-LMR Fertile		4-MHR Nonfertile		5-ATW Recycle
		1-Thru	Recycle	1-Thru	Recycle	1-Thru	Recycle	
Np-237	5.0	3.7	2.6	3.9	3.1	5.2	5.2	2.0
Pu-238	1.3	5.9	7.9	4.1	5.4	6.9	11.0	6.2
Pu-239	53.2	34.8	22.7	48.1	46.1	23.8	13.8	18.5
Pu-240	21.5	31.6	37.6	25.3	29.3	24.1	22.2	35.4
Pu-241	3.8	4.8	5.8	3.5	3.9	18.1	20.4	6.8
Pu-242	4.7	7.2	9.6	5.6	6.5	9.0	12.7	13.0
Am-241	9.0	7.6	6.7	6.8	5.5	5.7	4.5	5.1
Am-242M	0.0	0.6	0.7	0.4	0.5	0.2	0.1	0.5
Am-243	0.9	1.8	2.7	1.3	1.7	3.1	4.8	4.4
Cm-242	0.0	0.3	0.2	0.2	0.2	2.0	1.9	0.5
Cm-243	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1
Cm-244	0.1	0.7	1.4	0.4	0.7	1.2	2.5	3.7
Cm-245	0.0	0.1	0.3	0.0	0.1	0.2	0.3	1.0
Cm-246	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.6
Fissile Pu	57.0	39.6	28.5	51.7	50.0	41.9	34.2	25.3
Np-Chain	17.8	16.2	15.4	14.3	12.6	29.2	30.3	14.8

except for power peaking where the source computations are required.

The key performance parameters are compared in Table IV. The fissile content variations of the feed compositions impact the TRU inventory requirements. The highest inventory is obtained for the ATW recycle, the lowest inventory (30% lower) with the LWR feed. In general, higher fuel inventories are required when using the single-recycle feeds because they have lower fissile content (see Table III). The dispersion fuel fraction limit is violated for the highest burnup, lowest fissile fraction cases (LMR non-fertile recycle and ATW recycle). The reactivity loss rate decreases at higher TRU inventory, but the variations are small (only 1%Δk between the first strata cases). In addition, at high TRU inventory the discharge burnup is reduced proportionally since the residence time is conserved (a 25% variation between ATW recycle and LWR feed cases). However, the peak discharge fluence is nearly constant indicating that the burnup penalty cannot be recovered by extending fuel lifetime.

V. CONCLUSIONS

A variety of multiple strata options in which the LWR transuranics remain mixed (no separation of plutonium and minor actinides) were investigated. The utilization of first-strata fast or thermal spectrum reactors for partial burnup of TRU (before final transmutation in an ATW system) was considered. In addition, the impact of this irradiation on ATW performance was evaluated.

TABLE IV

Sodium-Cooled ATW Performance with Variation of External TRU Feed

Parameter		ATW Startup	LMR Nonfertile		LMR Fertile		GT-MHR Nonfertile		ATW Recycle
			1-Thru	Recycle	1-Thru	Recycle	1-Thru	Recycle	
Fissile plutonium fraction, %		57.0	39.6	28.5	51.7	50.0	41.9	34.2	25.3
BOEC Heavy metal inventory (kg)		2008	2388	2746	2056	2106	2193	2361	2885
Fuel pcle fraction (vol %)	Inner Zone	15.9	18.3	20.6	16.2	16.5	17.1	18.2	21.5
	Outer Zone	20.9	24.1	27.2	21.3	21.7	22.5	23.9	28.4
Multi-plication factor	BOEC	0.970	0.971	0.970	0.971	0.969	0.970	0.971	0.970
	EOEC	0.913	0.924	0.931	0.924	0.914	0.913	0.917	0.931
Burnup reactivity loss (%Δk)		5.70	4.72	3.94	5.63	5.56	5.66	5.37	3.88
Peak linear power (W/cm)		405	389	378	402	400	401	398	377
Discharge burnup (MWd/kg)		344	298	265	337	331	319	299	254
Peak fast fluence (10^{23} n/cm^2)		4.00	3.95	3.91	4.01	4.01	3.99	3.97	3.94

Results show that the multiple strata options can realize the primary goal of partial TRU consumption prior to the ATW campaign. For both the thermal and fast systems evaluated in this study, ~30% of the TRU mass was consumed in a once-through first strata irradiation. An additional 20-30% could be consumed with a single recycle in the first strata system. However, significant buildup of higher actinides (including the key radiotoxicity chain) was observed for thermal spectrum irradiation.

It appears quite difficult to achieve the second goal of improved ATW performance, with these concepts in which the plutonium and minor actinides remain mixed. Only small impacts on the burnup swing (1-2%Δk) were observed with significant penalties (~25%) in ATW discharge burnup. Furthermore, large decreases in the TRU fissile content will challenge the dispersion fuel particle fraction limits and likely require an alternative ATW fuel form. Other possible means to improve ATW performance will be investigated including separated TRU scenarios, heterogeneous ATW loading to exploit compositional variations, deep burnup in the first strata, and use of fertile fuel in the ATW final strata.

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